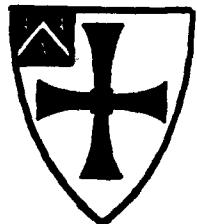


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REPORT ON THE INTERNATIONAL WORKSHOP
ON PROGRESS TOWARDS MOLECULAR SCALE
ELECTRONICS UNIVERSITY OF DURHAM

MARCH 25th - 28th 1990

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REPORT ON THE INTERNATIONAL WORKSHOP ON PROGRESS TOWARDS MOLECULAR SCALE
ELECTRONICS, UNIVERSITY OF DURHAM, MARCH 25TH - 28TH 1990

Introduction

The aim of the Workshop was to review recent theoretical and experimental progress towards the realisation of active electronic functions at, or near, the molecular scale. In order to achieve this the topics covered included:

- (a) the quantum mechanical modelling of electronically active molecules,
- (b) synthesis of target molecules and polymers and
- (c) physical characterisation of materials and processes.

While the focus was on the design, preparation and properties of molecules displaying electronic activity a number of related topics were covered:

- (a) electronic processes in biological systems,
- (b) fabrication of super-molecular structures and
- (c) appropriate computational schemes.

The oral presentations were scheduled with adequate time for discussion, which was particularly lively and stimulating and generally occupied much of the time allowed for programme breaks. A poster session enable most of those not giving papers to present their recent research results.

The full programme, abstracts of both oral and poster presentations and a list of participants with addresses, telephone and fax numbers is appended to this report.

Meeting summary

This summary is not intended to be a detailed account of the papers presented. It is intended to provide an overview which compensates, to some extent, for the large variation in detail in the abstracts provided by the participants.

Professor Mehring started the meeting by contrasting the potential of molecular scale electronics with the present status of silicon VLSI technology.

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He illustrated his discussion of molecular electronics with examples of the research being undertaken in the multidisciplinary Sonderforschungsbereich in Stuttgart. Progress was reported in the synthesis and characterisation of viologens and polyenes designed to exhibit switching and controlled charge separation. The theoretical analysis of these and target molecules, not yet synthesised, being conducted in parallel with the experimental studies was described.

The factors limiting the speed and power requirements of electronic devices were delineated by Professor Chiabrera. He drew analogies with natural systems showing that in general they function in a very different regime from that of silicon based devices. This discussion leads to the conclusion that new device and system concepts are likely to emerge in molecular scale electronics, a point reinforced by Professor Barker later in the meeting. The equivalence of information theory and thermodynamics at the molecular scale was emphasised.

The molecular design of metal-organic compounds for the photoexcitation of either energy transport or charge separation within the molecule was described by Professor Balzani. The use of quantum chemistry in the description of electron transfer along structured polymer chains was the focus of the contributions from Drs. Beratan and Joachim. Dr. Beratan considered photoexcited electron transport in molecules with energy level structures based on the highly efficient naturally occurring photoreaction centres of bacteria. Dr. Joachim's contribution detailed calculations of electron tunnelling probabilities of conjugated links containing aromatic and other moieties. He showed how such models could be extended to describe the behaviour of electrons in the electron tunnelling microscope. Dr. Beratan also highlighted the similarity in approach involved in modelling such conceptual molecular scale electronic processes and the calculation of the non-linear optical properties of molecules.

To round off the first day Professor Hong discussed charge separation processes in photosynthetic and visual membranes. The use of carefully chosen

equivalent circuit models was shown to be an important factor in gaining understanding of the fundamental properties of these highly complex systems. The possibility that such biologically derived materials could be used for molecular electronic purposes was also discussed.

The control of molecular and supramolecular structure was considered in the first session on the second day. Professor Feast emphasised the need for better control of synthesis and handling of polymers to ensure that meaningful measurements of properties can be made. The advances in synthetic methods that have facilitated the preparation of acetylene oligomers and polymers with well defined chain lengths and free from impurities were described. The utilisation of rigid molecules to build three dimensional arrays with alternation of units with chosen functionality was identified as of considerable significance for molecular scale electronics. This theme was continued by Dr Sagiv though his starting point was a functionalised solid surface rather than a single molecule. He also made the point that molecular scale electronics could, and probably will, involve materials other than purely organic compounds. Indeed heterogeneous structures could offer unique combinations of properties not otherwise achievable.

Professor Lahav described how the growth and properties of polar and chiral materials could be controlled by the adsorption and inclusion of specifically designed additives. The precise control of morphology with small additions of polymers with chosen pendant groups holds out the prospect of a novel route to supramolecular structures. This work also shows how centrosymmetric crystals can be tailored to give useful non-linear optical properties.

The use of the 'Durham' precursor route to polyacetylene in the production of novel FET based electronic devices was described by Dr. Friend. The novel device physics was emphasised. First, that defects in the conductive polymer chain produce a wider band-gap so that even an amorphous material has a low density of defect induced states in the band-gap. Secondly, charge concentration

in the FET gate region gives rise to mid-gap absorption and a large intrinsic electro-optic coefficient. These properties are unique to polymeric semiconductors and opens up prospects for application if the problem of the poor stability of polyacetylene can be solved. Dr. Horowitz discussed the semiconducting properties sexithiophene, a model material for the related polythiophenes.

Professor Barker gave a thought provoking talk on computer architectures which will probably be relevant for molecular electronics. Particular points were the need for finite state elements operating in a regime where quantum fluctuations are unimportant. Cellular automata were discussed as a universally computing structure which could be constructed from two-dimensional molecular arrays. The need for new computational concepts was emphasised as well as consideration of molecular structural reorganisation as a process on which to base molecular electronic devices. Finally the interface between nanotechnology and molecular electronics was discussed as a way towards hybrid structures and devices.

In the final session Dr. Welland discussed the mechanisms that give rise to contrast in the scanning tunnelling microscope (STM). The relevance of these to organic and biological materials and the problems encountered in obtaining STM images of these materials was described. Dr. Michel showed some stunning STM images, albeit computer enhanced, of biological samples and organic crystals. The latter in particular were much thicker than conventional wisdom suggests is the limit at which STM images can be obtained. Despite extensive discussion this remained a puzzle though the skill involved in obtaining the images remained unquestioned.

The poster session covered a variety of topics. Several focused on molecular design to achieve particular macroscopic physical properties, e.g. optical non-linearity, liquid crystallinity. This branch of molecular electronics covers materials which are either already used for technology or are

likely to be in the near future. Despite the emphasis on macroscopic, rather than microscopic, behaviour work in this area can have relevance to molecular scale electronics. For example a better understanding of the every-growing number of distinct liquid crystalline structures may well be important in the self-assembly of molecules in forms useful for molecular scale devices. It also has the important role of familiarising industrial scientist with organic materials and providing a more receptive audience for future developments in molecular scale electronics.

Conclusions

In my final summary I attempted to highlight the differences in approach to molecular scale electronics which had been presented during the meeting. On one hand the theoretical chemists are considering sophisticated quantum chemical calculations of molecular switches while on the other it was argued that the assembly of such elements was likely to pose insuperable problems and overall structure had to be considered from the outset. It can be argued that while much of the literature in the area shows great sophistication in the consideration of molecules it lacks it in the consideration of systems. There remains a wide gap between what has been achieved and the long term aim of molecular scale electronics. This meeting confirmed the view that we are only just starting to take the first steps down what is probably a long road.

This is particularly true in the experimental arena. The tools needed to manipulate and probe materials at the molecular scale are still of recent provenance and their use is still being explored. It is, however, clear that techniques such as scanning tunnelling, atomic force, near field optical and exciton microscopies provide the means of probing the properties of individual molecules and small molecular aggregates. This development has occurred at a time when progress in other disciplines is also converging on the molecular scale as indicated in Figure 1.

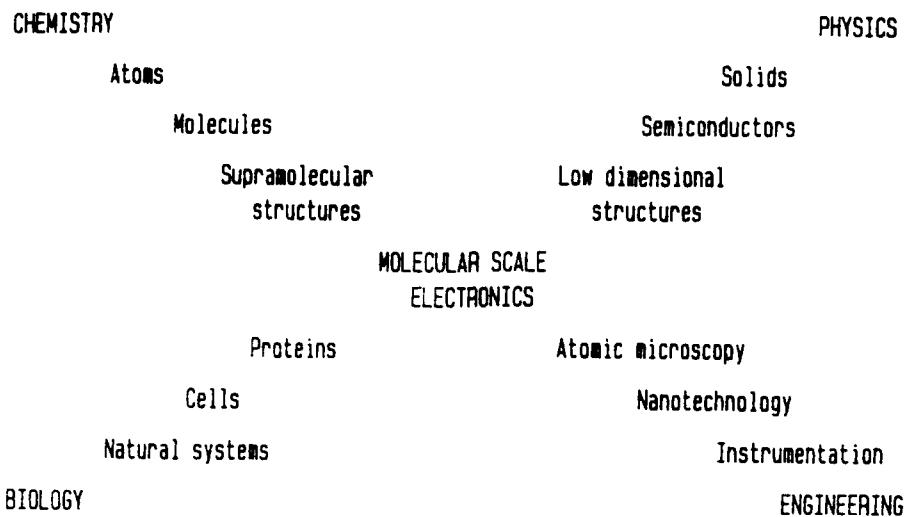


Figure 1. Convergence towards Molecular Scale Electronics

The papers presented at the workshop illustrated the wide range of potential routes to molecular scale electronics. However, it was possible to define two routes where the materials selected lead logically to possible device architectures. These are summarised in Table 1.

Table 1

Structures for Molecular Scale Electronics

Rigid - Solid State	Fluid - Solution
Rigid Molecules	Flexible molecules
Fixed arrays:	Dynamic arrays:
Two dimensional	Vesicles
Three dimensional	Membranes
Bi- or multi-stability:	Bi- and multi-stability:
Conformation	Molecular shape
Electronic state	Potentials
Transport:	Transport:
Electrons	Ions
Excitons	Molecules

Starting from small rigid molecular units the formation of ordered two or three dimensional arrays can be achieved by adsorption, the Langmuir-Blodgett method or direct synthesis. The transport of information could utilise either

electron or exciton transport. Long distance transport is not required if a cellular automation architecture is considered. For such a system energy input and timing could in principle be provided by a normally incident laser pulse train. The properties of such arrays are amenable to probing by the STM etc. Eventual devices would have a relatively small number of external connections either via STMs or nanoengineered structures. Data/programme structures could be either built in or impressed with light beams or a combination of both. The possibility exists of using structural changes either to assist carrier generation or as an active process in its own right.

A second approach could employ simplified analogues of ion-channels placed in a synthetic bilayer in an aqueous medium. The controlled opening of such channels would allow ions or possibly molecules to be used for information transport. Systems with one bit per carrier could then be implemented as ion transport would be less troubled by quantum effects. This is an analogue of natural synapses and because of the slow speed of the transport process would require parallel or multiply connected (neural net) architectures. Ion transduction would lead to electrochemical effects such as change in membrane potential. This could be coupled with redox processes to release charge carriers into conventional circuitry using nanoengineered interconnects. In such systems chemical reactions could be used to provide energy input and, via oscillatory reactions, timing. Asynchronous operation is, however, more likely in such systems.

While much basic research needs to be done to test these ideas, the means are now to hand in order to conduct the necessary microscopic scale experiments. In order to achieve progress expertise from a member of disciplines will have to be brought together. It is to be hoped that this will be synergistic and lead both to new ideas and more rapid progress than achievable by a less broadly based approach. To achieve the necessary breadth collaborations may need to span national as well as subject boundaries. Chemists have a vital role to play,

whatever approach is adopted, through their ability to synthesise materials with targeted properties to achieve self assembly into complex supramolecular structures and electronic and conformational properties of potential value for molecular scale electronics.

It is to be hoped that the Workshop stimulated those present and provided a contact point for future collaborations.

D. Bloor
May 1990

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**International Workshop on Progress Towards
Molecular Scale Electronics**

**University of Durham
Van Mildert College**

25th - 28th March 1990

**Sponsored by European Research Office of the
United States Army**

Organising Committee:

D. Bloor, University of Durham, U.K.
W. J. Feast, University of Durham, U.K.
M. C. Petty, University of Durham, U.K.
J. M. Lehn, Institute le Bel, France
J. R. Barker, University of Glasgow, U.K.
M. Mehring, University of Stuttgart, W. Germany
F. T. Hong, Wayne State University, U.S.A.
R. R. Burge, University of Syracuse, U.S.A.

International Workshop on Progress Towards
Molecular Scale Electronics

PROGRAMME

March, 25th

18.00 - 20.00 Registration and Buffet Van Mildert College

March, 26th

9.00 Welcome

9.05 Review of status of molecular electronics
Chairman: D. Bloor

9.05 - 10.05 M. Mehring, University of Stuttgart
Processes for Molecular Information Storage

10.05 - 11.05 A. Chiabrera, University of Genoa
Physical Limits of Computation in Information Molecular Processing

11.05 - Coffee -

11.45 Structure and function of molecules
Chairman: J. W. Feast

11.45 - 12.45 V. Balzani, University of Bologna
Design of Polynuclear Metal Complexes for Photoinduced Energy Migration and Charge Separation

12.45 - Lunch -

14.15 Theoretical models of molecular devices
Chairman: J-L Bredas

14.15 - 15.15 D. N. Beratan, Jet Propulsion Laboratory
Controlling Charge Transport and Polarization for Molecular Electronics

15.15 - 16.15 C. Joachim, CNRS Toulouse
Elastic Conductance Through a Single Molecule : Definition, Conformational and Interferencial Switches

16.15 - Tea -

17.00 Electronic processes in biological systems
Chairman: R. Pethig

17.00 - 18.00 F. T. Hong, Wayne State University
Electrochemical Analysis of Electronic Processes in Photobiological Systems

19.00 - Dinner -

20.30 Poster session and general discussion

March, 27th

9.00 Synthetic challenges, supramolecular assembly 1
Chairman: M. C. Petty

9.00 - 10.00 J. W. Feast, University of Durham
Precision and Control in the Synthesis of Polymers for
Molecular Electronics

10.00 - 11.00 J. Sagiv, Wiesmann Institute
Towards Organic-Inorganic Superlattice Structures via
Chemically-Controlled Self-Assembly

11.00 - Coffee -

11.45 Supramolecular assembly 2
Chairman: A. Barraud

11.45 - 12.45 M. Lahav, Wiesmann Institute
Strategies for the Design of Solids with Polar Structures

12.45 - Lunch -

14.15 Applications of molecular materials
Chairman: E. Ehrenfreund

14.15 - 15.15 R. Friend, University of Cambridge
MIS and MISFET Devices Fabricated with Conjugated Polymers

15.15 - 16.15 G. Horowitz, CNRS, Thiais
Conjugated Oligomers as Molecular Materials for Electronic
Devices

16.15 - Tea -

17.00 Computation for Molecular Electronics
Chairman: A. P. Monkman

17.00 - 18.00 J. R. Barker, University of Glasgow
Towards Computing at the Molecular Level

19.00 - Conference Dinner -

March, 28th

9.00 Characterization at the molecular level
Chairman: S. Roth

9.00 - 10.00 M. Welland, University of Cambridge
Image Contrast Mechanisms in STM and AFM

10.00 - 11.00 H. Michel, IBM Research Labs, Zurich
STM Investigation of Macromolecules

11.00 - Coffee -

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11.45 The Way Forward : Summary and Close of Meeting
 D. Bloor, University of Durham
 What does the Future hold for Molecular Scale Electronics?

12.45 - Lunch -

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MAIN PRESENTATION ABSTRACTS

Processes for Molecular Information Storage

Michael Mehring
2 Physikalisches Institut, Universitat Stuttgart, D-7000 Stuttgart 80, Fed. Rep.
Germany.

Elementary steps in molecular information storage as well as complex molecular structures, which are capable of storing information on a molecular level will be discussed. I will emphasize electron transfer processes in molecular chains and supermolecules. Polaron transfer storage will be proposed as a possibly bistable process. Coupling of different molecular units may result in processing information on a molecular level. Basic concepts and feasibility of the proposed units will be discussed.

Physical Limits of Computation in Information Molecular Processing

A. Chiabrera, E. Di Zitti and G. M. Bisio

Department of Biophysical and Electronic Engineering, University of Genoa, via
Opera Pia 11a, 16145 Genoa, Italy

Both conventional electronic circuits and molecular circuits must conform to the general law of thermodynamics because the functions they perform involve physical variables. The corresponding limits of computation can be evaluated and compared by measuring the information rate in entropic units. These latter results are preliminary and can be used for explaining simple paradoxes like the Maxwell's demon. Nevertheless, they could offer a general framework for the evaluation of the computational performances which can be achieved by molecular electronics.

Design of polynuclear metal complexes for photoinduced energy migration and charge separation

Vincenzo Balzani

"G. Ciamican", Department of Chemistry of the University and FRAE-CNR Institute, Bologna, Italy.

Several supramolecular transition metal complexes which contain from 2 to 7 metal centres and from 5 to 12 polypyridine-type ligands have been synthesized and their absorption spectra, luminescence spectra, luminescence lifetimes, oxidation and reduction properties, and interchromophoric energy transfer processes have been investigated. All the complexes examined exhibit very intense ligand-centred (LC) bands in the near UV region (ϵ up to 300000), and intense (ϵ up to 70000) and broad metal-to-ligand charge transfer (MLCT) bands which extend all over the visible region. All the complexes exhibit MLCT luminescence in the red or near infrared spectral region with lifetimes of the order of 1 μ s at 77 K and 100 ns in fluid solution at room temperature. The lowest (luminescent) excited state is populated with unitary efficiency, regardless of the excitation wavelength. In electrochemical experiments, the complexes show metal centred oxidation and ligand centered reduction processes. Most of the redox waves are reversible and can be assigned to specific metal(s) or ligand(s). The interaction between equivalent redox centres of the same complex is more or less weak, depending on the nature of the ligands.

These di-, tri-, tetra-, and hepta-nuclear complexes can be used as building blocks for the design of luminescent and redox-reactive species containing a higher number of metal centres. Because of their broad and strong absorption bands in the visible, relatively long luminescence lifetime, and rich redox behaviour, complexes of this type can prove useful as antenna components for photosensitization purposes (including electron or hole injection on semiconductors), luminescence probes and multielectron transfer photocatalysts.

Controlling charge transport and polarization for molecular electronics

David N. Beratan

Jet Propulsion Laboratory, California Institute of Technology, Pasadena, CA,
U.S.A.

A review of our recent theoretical work on problems in molecular electronics will be presented. This will include a description of the electron transfer shift register and synthetic target molecules, strategies for the design and interpretation of organic nonlinear optical chromophores, and strategies for mapping electron transfer pathways in complex macromolecules. The discussion will emphasize the connection between theory and experiment, and the role for theory in molecular electronics.

Elastic conductance through a single molecules : definition, conformational and interferencial switches

C. Joachim

Molecular Electronic Group, CEMES-LOE CNRS, 29 Rue J. Marvig, B.P. 4347, 31055 Toulouse Cedex, France

From an extension of the Landauer formula to intramolecular tunnelling processes, the elastic conductance of a molecule embedded between two nanoscopic size wires is defined. The conductance of ligands optimized for their functional properties in a mixed valence compound is calculated. After the example of a 3-level tight-binding system, example of an intramolecular conformational switch embedded between two gold wires (5 atoms in section) and of interferencial control of the conductance by substituted benzene will be discussed.

Electrochemical Analysis of Electronic Processes in Photobiological Systems

Felix T. Hong

**Department of Physiology, Gordon H. Scott Hall of Basic Medical Sciences, 540
East Canfield Avenue, Detroit, Michigan 48201.**

Photobiological systems such as photosynthetic and visual membranes are naturally occurring molecular optoelectronic devices. This presentation illustrates the usefulness of electrochemical analysis in understanding the factors that influence the time course of pulsed light induced electrical signals in model photobiological membranes. It was found experimentally that the electrical signals are subject to modulation by the load resistance, the electrical characteristics of the supporting inert structures, as well as the inherent photochemical characteristics of the photoactive components. Quantitative prediction of the time course of such signals can be done with the aid of equivalent circuit analysis, as is done in electronic devices constructed from inorganic electronic materials. However, chemical processes in a photobiological system also participate in the modulation of the signal time courses and therefore can be exploited in molecular electronics designs.

Precision and Control in the Synthesis of Polymers for Molecular Electronics

W. J. Feast

Interdisciplinary Research Centre in Polymer Science and Technology, Department of Chemistry, Durham University, South Road, Durham, DH1 3LE.

It is widely agreed that the targets for polymer synthesis during the next few years will be bespoke or speciality materials. Generally such materials will be needed in relatively small amounts rather than the high tonnages associated with the polymer industry over the last few decades. The properties of these new materials will have to be precisely defined, consequently absolutely reproducible syntheses are essential. This situation places an urgent requirement on synthetic chemists to develop methods for making novel polymers in which all its aspects of structure (connectivity, stereochemistry, conformation and molecular size and its distribution) can be precisely controlled.

These generalisations are nowhere more important than in the field of molecular electronics, and possibly nowhere more frequently ignored at some cost to the advancement of the subject.

Significant advances in polymer synthesis have been achieved in the last few years which have implications for the production of materials of interest in this field of research. Examples of well controlled syntheses conjugated polymers (polyenes, polyarenes, etc), stereoregular fluoropolymers, and other polymers of interest in molecular electronics will be discussed along with problems and limitations which need addressing.

Towards organic-inorganic superlattice structures via chemically-controlled self-assembly

Rivka Maoz and Jacob Sagiv

Department of Isotope Research, The Weizmann Institute of Science, Rehovot, Israel

Recognizing the need for novel materials, design concepts and methods of fabrication, Molecular Electronics has focused its attention on the wide, so far unexploited, potential of organics as materials of choice for the development of new types of functional elements and modes of operation not necessarily realizable through the use of simple inorganics only. On the other hand, remarkable recent advances in solid state electronics point to unforeseen new possibilities as regarding the miniaturization of electronic circuits, on the basis of some sort of inorganic materials that proved so successful in the present solid state technology.

This situation obviously generates strong conflicting arguments in favour of either the "organic" or the "inorganic" approach, while, in fact, one can find no real scientific reason for regarding these two sets of materials as mutually exclusive. We thus believe that a more comprehensive approach to "molecular scale electronics" should consider any feasible combination of materials and modes of assembly that may lead to useful electronic components of molecular dimensions.

In this lecture we wish to present recent experimental data demonstrating the controlled self-assembly of some novel solid-supported layered organizations made of both organic and inorganic constituents. Such organic-inorganic supramolecular assemblies can be synthesized by judiciously combining well known synthetic methods and principles of organic, inorganic and surface chemistry. The present preliminary results hold promise with regard to the possible realization of a new class of artificial superlattice structures, in which organic and inorganic components are combined and optimally integrated, so as to take full advantage of desirable specific properties of each of them.

Strategies for the Design of Solids with Polar Structures

M. Lahav and L. Leiserowitz

Department of Structural Chemistry, Weizmann Institute of Science, Rehovot 76100, Israel.

Solids of a non-centrosymmetric packing arrangement display generally unique physical properties such as piezo and pyroelectricity or optical frequency doubling. In many instances, however, their preparation is prevented by the preferential formation of a thermodynamically more stable centrosymmetric counterpart.

In this lecture we shall describe three different strategies for the preparation of polar solids. This will include: (i) control of polymorphism with assistance of tailor-made auxiliaries, (ii) reduction in symmetry in solid solutions and (iii) a new class of amphiphiles forming Z-type LB films.

1. E. Staar et al., Advanced Materials, in press.
2. I. Weissbuch et al., Chemistry of Materials, 1, 114-118 (1989).
3. R. Popovitz-Biro et al., J. Amer. Chem. Soc. in press.

MIS and MISFET Devices Fabricated with Conjugated Polymers

R. H. Friend

Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge,
CB3 OHE, UK.

The semiconductor physics of conjugated polymers is very different from that of conventional inorganic semiconductors, and it is in the novelty of the physics that has been investigated over the past decade or so that possible applications in technology have been identified. Two of the distinguishing aspects of the physics of conjugated polymers are: firstly, that charge transport is inherently anisotropic, with ready motion of charge along chains, and slower transport between chains, and secondly, that charge stored on the polymer chain brings about a local reorganisation of the π -bonding in the vicinity of the charge.

We have been able to fabricate various semiconductor devices using Durham-route polyacetylene as the active semiconductor. The Metal-Insulator-Semiconductor (MIS) structure and the MISFET are of particular interest because, with the formation of charge accumulation, inversion and depletion layers, charge can be injected into and removed from the semiconductor without the need for chemical doping. Solitons are the lowest energy electronic excitations of polyacetylene, and we show here the various spectroscopic indications that the active charges in these structures have the character of charged solitons. Image Contrast Mechanisms in Scanning Tunnelling Microscopy and Atomic Force Microscopy

Conjugated Oligomers as Molecular Materials for Electronic Devices

G. Horowitz and F. Garnier

Laboratoire des Materiaux Moléculaires, C.N.R.S., 2 rue Henri-Dunant, 94320
Thiais, France

Conjugated oligomers, such as thiophene derivatives, can be synthesized as well defined easily processable compounds. Their structural properties and the current-voltage characteristics of metal-semiconductor junctions fabricated from oligomers of variable chain length show that, even with a relatively short length of 6 thiophene units, these compounds possess electronic properties similar to those of their parent polymers, together with greatly enhanced semiconducting properties. This last result must be related to the presence of many less structural defects in the oligomers when compared to the polymers. The characteristics of thin film transistors fabricated from these oligomers confirm the potential interest of these molecular materials for use as active components in electronics. This first step opens the possibility of scaling down these devices.

Towards computing at the molecular level

J. R. Barker

Nanoelectronics Research Centre, Department of Electronics and Electrical Engineering, University of Glasgow, Glasgow, G12 8QQ.

The prospect of chemically or biologically engineered molecular structures which might support information processing functions has enticed workers for many years. Many of the top-down theoretical studies of the "molecular computing architectures" which might be realisable have ignored the practical constraints of dealing with molecular materials but instead have emphasised the possible massive parallelism or storage capacity and even the possibility of a self-organised fabrication of a supramolecular computer. At the other viewpoint, the bottom-up approach has sought to follow the traditional route of "wires", "circuit components", "switches" and "gates" to "circuits", essentially analogous to the development of semiconductor technology. In the present paper we argue that the practical constraints force us to much less ambitious structures which emphasise the need for stable materials which can be structured into very simple arrangements capable of simple interfacing to the outside world. We argue that the molecule-centred approach, essentially a chemistry driven approach is required which uses the designed-in functionality of a particular molecule to represent in a single entity the equivalent of a high level logic structure such as a finite-state machine rather than a sub-component of a "molecular transistor" or "molecular logic gate". The interconnection and interaction of such molecules would provide data paths and local switching functions.

The following issues are discussed: (a) the appropriate architectural targets and the implication for molecular design, the discussion will be based mainly on ideas from the field of cellular automata; (b) the one bit per "carrier" limit, its molecular representation and how it alters logic design; (c) a novel method for the chemical synthesis of an interface between a solid state nanocircuit and an array of organic molecules organised on sub-micron scales (this is the subject of a current experimental investigation at Glasgow involving the Departments of Biochemistry, Cell Biology and Electronic and Electrical Engineering) and (d) possible mechanical molecular computing structures.

Image Contrast Mechanisms in Scanning Tunnelling Microscopy and Atomic Force Microscopy

M. B. Welland

Cambridge University, Engineering Department, Trumpington Place, Cambridge.

This presentation will review the basic mechanisms for producing image contrast in the scanning tunnelling microscope (STM) and the atomic force microscope (AFM). The bulk of the talk will concentrate on the STM since there are a variety of mechanisms which can produce image contrast which are particularly relevant to imaging molecular species on a conducting substrate. These will include dipole image effects, density of state effects, small scale contact, asymmetric tunnel barriers and contrast due to isolated charge. The latter effect may be particularly relevant to molecular electronics since the STM is very sensitive to isolated charge so that the prospect of imaging a charged carrier as it moves through a molecular device becomes possible.

The contrast mechanisms in AFM are perhaps not as clear cut as in STM as the interactions between tip and surface are more difficult to model. The resolution depends upon which part of the force curve the AFM is operating in and also on elastic interactions between tip and surface which may result in atomic diffusion or in an extreme case defect generation.

STM Investigation of Macromolecules

B. Michel

IBM Research Division, Zurich Research Laboratory, CH-8803 Rüschlikon

A central issue of STM investigations of biological molecules is the electron transfer mechanism. Most biological substances are considered to be bad conductors or even insulators in the bulk state. The first STM images of macromolecules of tens of nm in size came therefore rather as a surprise. This in view of the large electron transfer rate of 10^{10} electrons per second through a cross section of a few square Ångstroms. Examples from all major classes of biological molecules have been imaged since : DNA, RNA, protein, carbohydrates, and lipid molecules, as well as supramolecular structures like Langmuir-Blodgett films, biological membranes, viruses and small cells.

The complicated electronic and chemical structure of biological molecules makes it difficult to differentiate experimentally between intrinsic and extrinsic electron transfer. We therefore performed STM experiments on the paraffins or n-alkanes, the most simple organic macromolecules. STM imaging of n-heptadecane, n-octadecane and n-hexatriacontane has been possible for structures with a thickness of up to 100 nm and we also observed melting, induced modification, and recrystallization processes. Since alkanes are hydrophobic and non-polar, we can exclude electron transfer hypotheses based on surface conduction and ion-conduction.

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POSTER PRESENTATION ABSTRACTS

Synthesis of molecules for molecular arrays : model systems for molecular electronics

R. Bonnett, M. Rogers-Evans, S. Pugh

Departments of Chemistry and Physics Queen Mary Westfield College
London, U.K.

D. Bloor

Applied Physics Group, University of Durham, U.K.

Many routes are open for the development of molecular electronic devices. One possibility is the assembly of regular arrays of active molecules that could function as a cellular automaton. This approach requires the synthesis of molecules that will form regular two-dimensional arrays that either possess an appropriate functionality or can act as a host matrix for active molecules. Materials meeting these requirements are being synthesised. These will be formed into arrays by either adsorption or via the Langmuir-Blodgett film technique for eventual study by STM and other molecular scale resolution techniques.

Columnar Liquid Crystals from Poly(di-n-Alkylsilanes)

P. Weber, D. Guillon and A. Skoulios

Institute de Physique et Chimie des Materiaux de Strasbourg Groupe des Materiaux Organiques, I.C.S. Strasbourg, France

R. D. Miller

IBM Almaden Research Center, San Jose, California, USA

A homologous series of poly(dialkylsilanes), with alkyl side-chains from butyl to tetradecyl, was studied using polarising optical microscopy, differential scanning calorimetry, and x-ray diffraction. It was shown that these polymers undergo a first order transition from a low temperature crystalline to a high temperature columnar mesomorphic phase. The transition temperatures measured range from 9 to 82°C. As the length of the side-chains is increased, the transition temperature initially drops drastically and then rises again. These two regimes are connected to the relative part played in the phase transition by the aliphatic side-chains and the polymer backbone. Regarding the enthalpies, two regimes were also detected. Starting from the butyl polymer the enthalpy first grows more and more rapidly and beyond a chain length of about ten methylene groups it then grows linearly. From the Y-intercept of the corresponding curve it was inferred that the transition into the mesomorphic phase is followed by a stabilization of the backbone conformation. The structure of the columnar mesophase was described as a two-dimensional hexagonal packing of columns consisting of straight polymer backbones surrounded by alkyl chains in a disordered conformation. From the slope of the squared intercolumnar distance as a function of the number of methylene groups per silicon atom, and using the known value of the volume of one methylene group in the liquid state, the stacking period of the silicon atoms along the columnar axis was found to be equal to 1.68 Å. This relatively small value indicates a coiled structure involving quite an amount of gauche conformers. The Y-intercept of the corresponding straight line, related to the volume of one silylene repeat unit deprived of all its methylene groups, suggests a great compacity of the atomic packing near the silicon backbone.

Electrochemical Characterisation of Redox Polymers

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Normal pulse voltammetric (NPV) studies are reported for electrodeposited films of the model redox polymer, polyvinylferrocene. Of significance has been the appearance of a peak response in the current-voltage profile, instead of the current saturation effect which is normally observed in NPV studies on redox species in solution. The characteristics of the current peak are controlled by the effect of the film/solution interface in limiting charge transfer across the electrode/film interface. A theoretical analysis of the profile of the current peak provides good estimates of the electron transfer rate constant ($6.5 \times 10^{-5} \text{ cm s}^{-1}$) at the electrode/film interface, the charge diffusion coefficient ($1.35 \times 10^{-8} \text{ cm}^2 \text{s}^{-1}$) in the film bulk, the film resistivity ($10^4 - 10^5 \text{ ohm cm}$), as well as the physical integrity of the film. As such, NPV measurements provide a valuable method for determining information of importance in assessing the possible use of redox polymer films in the development of molecular electronic devices or sensors.

Theoretical Investigation of High Second-Order Polarizabilities in Push-Pull Polyenes

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Lehn and co-workers have recently reported the synthesis of push-pull polyene molecules where the donor is a benzodithia group and the acceptor, an aldehyde group. Polyene segments contain from 1 up to 8 double bonds. EFISHG measurements by Barzoukas et al indicate that these molecules display among the largest $\mu\beta$ values ever observed.

In this contribution, we present Hartree-Fock ab initio 3-STG calculations on the geometric structures, molecular dipole moments, and second-order polarizabilities (β) of benzodithiopolyenoils, as a function of the length of the polyenic segment. It is found that the dipole moment has a very weak dependence on the extension of the polyene segment ($\mu : n^{0.1}$) while β undergoes a strong evolution ($\beta : n^{2.8}$). The microscopic origin of these results is discussed in detail.

Polynuclear Metal Complexes as Antenna-Sensitizer Molecular Devices

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The photophysical behaviour of polynuclear metal complexes is characterized by the widespread occurrence of intercomponent energy and electron transfer processes. In suitably designed polynuclear complexes, intercomponent energy can be used to achieve "antenna effects", whereby the light energy absorbed by a number of chromophoric components is conveyed to a common acceptor, where it can be used for a variety of purposes.

In this contribution, we describe an application of the antenna effect in the field of dye-sensitization of semiconductors. This technique, developed to drive photoinduced processes on wide-bandgap semiconductors using sub-bandgap excitation, has its main drawback in the poor light harvesting efficiency of the

adsorbed dye. The idea is to replace a conventional dye with an antenna-sensitizer molecular device (i.e., a supermolecule consisting of covalently bound antenna and sensitizer components). The sequence of events required to obtain the desired function is: (i) efficient light absorption by the antenna components, (ii) efficient energy transfer from the antenna components to the sensitizer component, (iii) efficient charge injection from the sensitizer into the semiconductor.

This strategy has been tested using the cyano-bridged trinuclear complex $[\text{Ru}(\text{bpy})_2(\text{CN})_2\text{Ru}(\text{bpy}(\text{COO})_2)_2]^{2-}$ ($\text{bpy} = 2,2'\text{-bipyridine}$ and $\text{bpy}(\text{COO})_2^{2-} = 4,4'\text{-dicarboxy}, 2,2'\text{-bipyridine}$) as an antenna-sensitizer device on TiO_2 . The design of this V-shaped molecule involves the terminal $\text{Ru}(\text{bpy})_2(\text{CN})_2^-$ units as potential antenna components and the central $-\text{Ru}(\text{bpy}(\text{COO})_2)_2^{2-}$ unit as the potential sensitizer. Adsorption capability onto TiO_2 is provided by the carboxylate groups on the sensitizer component. Photophysical measurements in solution and photo-current spectra on TiO_2 electrodes demonstrate that this complex performs indeed according to mechanism (i)-(iii) and constitutes a convenient antenna-sensitizer device for sensitization of TiO_2 . Devices with the same basic molecular design and more extended antenna units are conceivable.

Electroactive Langmuir-Blodgett Films of Tetrathiafulvalence Derivatives

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Mono-substituted tetrathiafulvalene derivatives form stable, high-quality Langmuir-Blodgett (LB) films which are highly conducting after iodine doping ($\sigma_{\text{max}} = 1 \text{ S cm}^{-1}$). The molecular arrangements within these films have been studied by a range of techniques. Alternate-layer LB films of TTF and TCNQ derivatives, both substituted with one long chain, have been fabricated, and shown to be semi-conducting ($\sigma_{\text{rt}} = 5 \times 10^{-3} \text{ S cm}^{-1}$).

Transduction of chemical interaction into electronic signals

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Synthetic receptor molecules that selectively bind charged guests can store chemical information. The transduction of this information into electronic signals connects the chemical and electronic domains. Field effect transistors (FETs) are attractive transducing elements because these microdevices are able to register and amplify chemical changes at the gate oxide surface of the semiconductor chip.

Integration of molecular receptors and field effect transistors into one chemical system gives a device that can communicate changes of substrate activities in aqueous solution. Simulations of a system in which the receptor molecules are directly attached to the FET gate oxide indicate serious limitations with respect to sensitivity, dynamic range and extreme requirements for complex stability. Therefore we have concentrated on the integration of covalently attached thin membranes.

The problem of the thermodynamically ill-defined oxide-membrane interface has been solved by applying a covalently linked hydrophilic polyhydroxyethylmethacrylate (polHE-MA) gel between the sensing membrane and the silylated gate oxide. A buffered aqueous electrolyte solution in the hydrogel renders the

surface potential at the gate oxide constant via the dissociation equilibrium of the residual silanol groups. The subsequent attachment of a polysiloxane membrane that has the required dielectric constant, glass transition temperature T_g , and receptor molecule, provides a stable chemical system that transduces the complexation of cationic species into electronic signals (CHEMFET).

The response to changing K concentrations in a solution of 0.1 M NaCl is fast (< 1 sec) and linear in the concentration range of 10^{-5} - 1.0 M (55-58 mV/decade). A reference FET (REFET) based on the same technology is obtained when the intrinsic sensitivity to changes in ion concentration is eliminated by the addition of 2.10^{-5} mol g⁻¹ of didodecyldimethyl-ammonium bromide to the ACE membrane. Differential measurements with a REFET/CHEMFET combination showed excellent linear K response over long period of time.

All chemical reactions used are compatible with planar IC technology and allow fabrication on wafer scale.

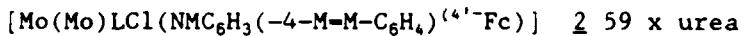
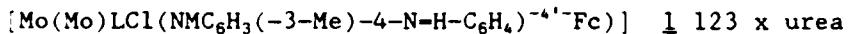
Molybdenum and Tungsten Mononitrosyl Complexes which exhibit Non-Linear Optic Properties

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A range of compounds has been prepared in which an electron withdrawing ($M(MO)LX(Y^-)$) ($M+M_0$, X=Cl or I; M=W, X=Cl; Y=Mo,NH; L=hydrotris(3,5-dimethylpyrazol-1-yl)borate) moiety is linked to an OMe or ferrocenyl ($Fc=(C_5H_5)Fe(C_5H_4^-)$) donor group by a polarisable aryl, stilbenyl or 4,4'-diazadiaryl bridging group. The electrochemical properties of the compounds have been determined by cyclic voltammetry and are in accord with their formulations. Three of the compounds exhibit substantial second harmonic generation (SHG) as follows:-



The X-ray crystal structure of 1 shows that the Mo (acceptor)-Ferrocenylcarbon (donor) axis of the molecule is oriented at 57° to the crystallographic 2-fold axis (b in $P2_1$). Powder X-ray diffraction studies indicate that 2 has a different structure but that the tungsten complex, 3, is isostructural with its molybdenum containing analogue, 1. Thus the difference in the observed SHG for 1 and 3 may be attributed to the electronic differences between Mo and W and to the poorer acceptor properties of W as compared to Mo.

Fluorescence Quenching in Tetraphenylporphyrin Derivatives carrying Peripheral Molybdenum Mononitrosyl Redox Centres

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A series of complexes has been prepared in which the triphenylperphycin moiety (TPPH_2) is linked via an aryl bridge to the redox active ($\text{Mo}(\text{Mo})\text{LX}(\text{Y}-)$) ($\text{X} = \text{I}, \text{Cl}$ and $\text{Y} = \text{para-O}$ or MH ; $\text{X} = \text{Cl}$, $\text{Y} = \text{ortho}$ and meta-O or MH ; $\text{X} = \text{OH}$, NHCH(Me)Ph and $\text{Y} = \text{para-O}$; $\text{L} = \text{hydrotris(3,5-dimethylpyrazol-1-yl)borate}$) moiety. The fluorescence of the tetraphenyl-porphyrin moiety is quenched to an extent which decreases with increasingly cathodic redirection potentials for the ($\text{Mo}(\text{Mo})\text{LX}(\text{para-O})$) moiety. Time resolved spectral studies by M. George, M. Policekoff, J. Kelly, and J. Turner have shown that the yield of the triplet state is $[\text{Mo}(\text{Mo})\text{L Cl(O-para-C}_6\text{H}_4\text{-TPPH}_2)]$ corresponds with the extent of fluorescence quenching and control shows that the fluorescence quenching is not due to a heavy atom effect. E.p.r. and infra red studies are being carried out in an attempt to demonstrate unequivocally that the fluorescence quenching arises from charge separation resulting from photo-induced electron transfer from porphyrin to molybdenum.

Amphiphilic Dyes for Second Harmonic Generation

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In this poster the synthesis of derivatives of the amphiphilic nitrophenylhydrazone and styrylpyridinium compounds, shown in the publication of Lupo et al^[1] is reported. All compounds shown form LB-monolayers at the gas-water interface and can be transferred to solid supports. These deposited monolayers are characterized via UV/Vis - and SHG-spectroscopy to determine the average tilt angle of the chromophores to the substrate and the nonlinear-optical properties of the compounds.

The nonlinear-optical results of the SGH-measurements can be summarized as follows:

- (a) Introduction of alkyloxy-substituents of the donor side meta to the bridge send to decrease molecular nonlinearity while ortho or para substituting enhances β due to the possibility of mesomeric structures.
- (b) Addition of a second nitro group at the acceptor moiety of the molecule increases β , but at the cost of increasing the apparent average tilt of the chromophore from the normal.
- (c) Somewhat surprising is the result that increasing the length of the conjugated π -system by one $-\text{CH}=\text{CH}-$ double bond has no apparent effect on β .
- (d) The thioether group compared to the alkyloxy substituent is found to increase β significantly without shifting the absorption maximum closer into resonance.

[1] D. Lupo, W. Prob, U. Scheunemann, A. Laschewsky, I. Ledouxx, H. Ringsdorf, J. Opt. Soc. Am., B, Vol. 5, 300 (1988).

Doping Induced and Photoinduced Absorption In β -Carotene

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By doping β -carotene with iodine, we demonstrate that a charged defect is produced. This defect has characteristics very similar to charged solitons (S^+) in *trans*-(CH)_x. It has only one mid-gap electronic transition peaked at approximately 1.3 eV, and a set of doping induced infrared active vibrations (IRAV), which absorb in the range 900–1600 cm⁻¹. These results indicate that a soliton-like defect can be generated in a polyene chain of only 22 carbons.

To compare further β -carotene with *trans*-(CH)_x we have measured the intra-gap absorption under the illumination of visible light with photon energy above the $\pi - \pi^*$ optical transition. Using this method (the "photoinduced absorption" method) we observed several mid-gap electronic transitions in the range 0.8 – 1.3 eV, but no photoinduced IRAV. These observations are markedly dissimilar to the photoinduced absorption spectrum of *trans*-(CH)_x, where a single mid-gap electronic transition accompanied by a set of IRAV were observed. The results of the photoinduced absorption measurements thus indicate that charge separation is inhibited in β -carotene, due probably to its short extent. The observed mid-gap transitions may be characteristic of few long lived triplet excitons, escaping the fast recombination of electron-hole pairs produced by the impinging light.

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